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### **Contaminant Concentrations in Juvenile Fall Chinook Salmon** from Columbia River Hatcheries

Lyndal L. Johnson, Maryjean L. Willis, O. Paul Olson, Ronald W. Pearce, CATHERINE A. SLOAN, AND GINA M. YLITALO

National Oceanic and Atmospheric Administration. Northwest Fisheries Science Center. Environmental Conservation Division, 2725 Montlake Boulevard East, Seattle, Washington 98112, USA

Abstract.—Previous studies have reported elevated chemical contaminant concentrations in out-migrant juvenile salmon from the lower Columbia River. Hatchery rearing is a potential exposure pathway, as contaminants have been measured in hatchery fish and feed from other regions. In this study, we analyzed for polychlorinated biphenyls (PCBs), organochlorine pesticides including dichloro-diphenyl-trichloroethanes (DDTs), polycyclic aromatic hydrocarbons (PAHs), and polybrominated diphenyl ethers (PBDEs) in juvenile fall Chinook salmon Oncorhynchus tshawytscha and feed from eight hatcheries (Big Creek Hatchery, Oregon Department of Fish and Wildlife; Elochoman, Cowlitz, Washougal, Klickitat, and Priest Rapids hatcheries, Washington Department of Fish and Wildlife; Little White Salmon National Fish Hatchery; and Spring Creek National Fish Hatchery) that release fish into the lower Columbia River. In feed samples, the mean concentrations of summed PCBs, summed DDTs, and summed PAHs were 14, 27, and 370 ng/g wet weight, respectively. In Chinook salmon bodies, mean concentrations of summed PCBs, summed DDTs, and summed PAHs were 17, 9.0, and 30 ng/g wet weight, respectively; metabolites of PAHs were also detected in Chinook salmon bile. Other organochlorine pesticides were detected at low levels (<5 ng/g wet weight) in feed and Chinook salmon from all hatcheries. Concentrations of PBDEs in feed and fish from all hatcheries were low (<3 and <1 ng/g wet weight, respectively). Contaminant exposure levels in hatchery Chinook salmon were generally below those associated with adverse effects on salmon health and also lower than those in fieldcollected juvenile fall Chinook salmon of hatchery origin from the lower Columbia River, suggesting that the river is a more important source of contamination than are the hatcheries.

Recent studies show that threatened and endangered juvenile fall Chinook salmon Oncorhynchus tshawytscha (Myers et al. 1998) that rear and feed in the lower Columbia River are accumulating persistent organic pollutants (POPs) at concentrations that could potentially reduce their survival (Johnson et al. 2007a, 2007b; LCREP 2007). Thus, identifying contaminant sources and reducing exposure are priorities for the recovery of these stocks (LCREP 2007; USEPA 2009). The listed stocks include fish of hatchery origin, which cannot always be reliably distinguished from wild fish through genetic analyses or hatchery marking (Myers et al. 1998, 2006). This raises the possibility that contaminants absorbed during hatchery rearing may contribute to body burdens in juvenile fall Chinook salmon from the lower Columbia River. Concern about this issue has been heightened by reports of chemical contamination in farmed and hatchery salmon throughout Europe and North America (Easton et al. 2002; Parkins 2003; Hites et al. 2004a, 2004b), including salmon from some Pacific Northwest hatcheries (Johnson et al. 2007a). If such contaminants are

present in hatchery-reared salmon released into the lower Columbia River, these fish could be contributing to the average contaminant body burdens for the juvenile salmon populations in the area, reducing the viability of hatchery stocks and acting as a source of contaminants for fish-eating predators.

In the present study, we measured concentrations of several classes of POPs in subvearling fall Chinook salmon from eight Columbia River hatcheries (Figure 1) before their release into the river in late May or early June. These hatcheries are thought to be major contributors to Chinook salmon populations in the lower Columbia River based on release data (CBR 2008) and genetic analyses of previously collected juvenile fall Chinook salmon collected from lower Columbia River sites (Johnson et al. 2007b). Altogether, the sampled hatcheries accounted for 74% of subyearling fall Chinook salmon released into the lower Columbia River in 2005, the year in which the present study was conducted (CBR 2008), and for about 79% of subyearling fall Chinook salmon released into the Columbia River in 2008 (CBR 2008).

The contaminants measured in Chinook salmon and hatchery feed included (1) polychlorinated biphenyls (PCBs), industrial chemicals that were banned in the USA in the 1970s but that are still common in urban

<sup>\*</sup> Corresponding author: lyndal.l.johnson@noaa.gov Received November 25, 2008; accepted May 2, 2009 Published online November 5, 2009

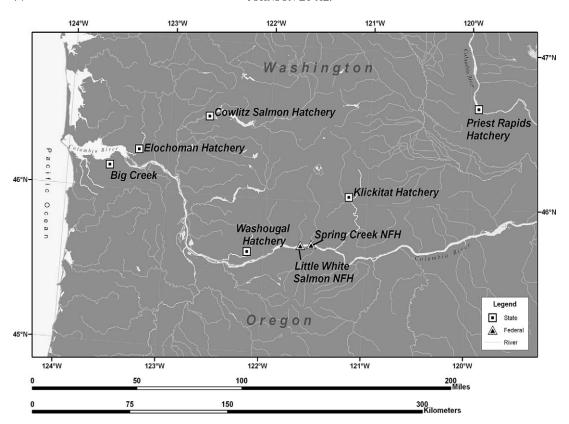


FIGURE 1.—Locations of Columbia River hatcheries in (Washington and Oregon) that were sampled as part of this study (NFH = National Fish Hatchery).

waterways (ATSDR 2000); (2) polybrominated diphenyl ethers (PBDEs), which are extensively used as flame retardants (ATSDR 2004); (3) dichloro-diphenyl-trichloroethanes (DDTs), insecticides that were banned in the United States in 1972 but that are still present in many agricultural areas (ATSDR 2002); and (4) polycyclic aromatic hydrocarbons (PAHs), which are derived from crude oils and various petroleum products (ATSDR 1995). These chemicals are associated with reproductive and developmental defects, immunosuppression, cancer, poor growth, and metabolic disorders in juvenile salmon and other fish (e.g., Incardona et al. 2005; Meador et al. 2006, 2008; Johnson et al. 2008). We also determined lipid content in Chinook salmon bodies because of its effect on contaminant uptake and toxicity (Elskus et al. 2005). The tissue concentration of a toxic lipophilic chemical is directly related to the amount of lipid in an organism, and when lipid content is high a higher proportion of the compound is associated with the lipid and is unavailable to cause toxicity (Lassiter and Hallam 1990; van Wezel et al. 1995).

Our objectives were to (1) determine whether POPs

were present in Columbia River hatchery Chinook salmon and commercial feed at concentrations that could affect fish health or pose a threat to piscivorous wildlife and (2) evaluate the potential contribution of hatchery feed to contaminant body burdens of outmigrant fall Chinook salmon by comparing contaminant levels in hatchery fish with previously determined contaminant concentrations in juvenile fall Chinook salmon of hatchery origin from several sites the lower Columbia River (LCREP 2007).

#### Methods

Fish Collection

In May 2005, bodies of juvenile subyearling Chinook salmon were obtained from eight hatcheries along the Columbia River (Big Creek Hatchery, Elochoman Hatchery, Cowlitz Hatchery, Washougal Hatchery, Little White Salmon National Fish Hatchery [NFH], Spring Creek NFH, Klickitat Hatchery, and Priest Rapids Hatchery; Figure 1). Big Creek Hatchery is operated by the Oregon Department of Fish and Wildlife; Elochoman, Cowlitz, Washougal, Klickitat, and Priest Rapids hatcheries are operated by the

Washington Department of Fish and Wildlife; and Little White Salmon NFH and Spring Creek NFH are operated by the U.S. Fish and Wildlife Service. All collections were performed shortly before the release dates for juvenile subyearling Chinook salmon so that contaminant concentrations would be typical of those in juvenile fish when they entered the lower Columbia River.

In addition to fish, two feed samples of approximately 10 g each were obtained from hatchery personnel at the time the fish were sampled. The feed samples were taken from the type and lot of feed that the fish were currently consuming. The feeds came from various commercial suppliers based in the Pacific Northwest. The feed samples were placed in 118-mL (4-oz) glass jars rinsed with isopropyl alcohol and were stored in a cooler with dry ice for transport back to the Northwest Fisheries Science Center (NWFSC) laboratory in Seattle. At the laboratory, feed samples were stored at  $-20^{\circ}$ C until chemical analyses were performed.

Approximately 30-40 individual fish at each hatchery were collected for necropsy. Fish were measured (to the nearest 1 mm), weighed (to the nearest 0.1 g), and then euthanized by a blow to the head. For each fish, bile was collected (when present) and the individual bile samples were composited into 4-mL glass vials containing glass 250-μL inserts. Bile samples from 10 to 20 individual fish per hatchery were pooled to obtain 3-5 µL of bile for measurement of PAH metabolites. The stomach and gastrointestinal tract were then removed, and stomach contents were extracted. These internal organs were then placed back into the visceral cavity of the carcass so that the tissue analyzed would consist of the whole body minus stomach contents. The carcasses containing the internal organs were individually wrapped in foil and labeled. Both body and bile samples were placed in a cooler with dry ice for transport back to the NWFSC laboratory in Seattle. At the laboratory, bile and body samples were stored at -80°C until chemical analyses were performed.

#### Sample Analyses

Lipid determination.—For lipid and chemical analyses, individual Chinook salmon bodies (carcass plus internal organs) from each hatchery were combined to produce composite samples consisting of 10 fish each. The amount of total, nonvolatile, extractable lipid (reported as percent lipid) in the body composites and feed samples was determined by gravimetric analysis as described in Sloan et al. (2004). Lipid classes were determined using thin-layer chromatography—flame ionization detection (TLC—FID) with Iatroscan analysis

as described by Ylitalo et al. (2005). The TLC-FID analysis also provided an estimate of percent lipid content, which was compared with the values determined gravimetrically. Lipid measurements obtained by gravimetric analysis were used to normalize body contaminant concentrations for lipid content. With the exception of one feed sample from Big Creek Hatchery, lipids in all samples were measured by both the gravimetric and TLC-FID methods; in the feed sample from Big Creek Hatchery, lipid content was measured by TLC-FID only.

Chemical contaminants in feed and body samples.— Body composite and feed samples were analyzed by gas chromatography-mass spectrometry (GC-MS) for PCB congeners, DDTs, DDT isomers, and other organochlorine (OC) pesticides (hexachlorocyclohexanes [HCHs], hexachlorobenzene [HCB], chlordanes, aldrin, dieldrin, mirex, and endosulfans) as described by Sloan et al. (2005). The PBDEs were measured similarly and concurrently in the GC-MS analyses. A total of 47 individual PCB congeners were measured (International Union of Pure and Applied Chemistry [IUPAC] numbers 17, 18, 28, 31, 33, 44, 49, 52, 66, 70, 74, 82, 87, 95, 99, 101/90, 105, 110, 118, 128, 138/ 163/164, 149, 151, 153/132, 156, 158, 170/190, 171, 177, 180, 183, 187/159/182, 191, 194, 195, 199, 205, 206, 208, and 209). The HCHs measured included  $\alpha$ -HCH, β-HCH, and γ-HCH (lindane). Dichloro-diphenyl-trichloroethanes measured included p,p'-DDT, p,p'-DDE, p,p'-DDD, o,p'-DDD, o,p'-DDE, and o,p'-DDT. Chlordanes and related compounds measured included heptachlor, heptachlor epoxide, γ-chlordane, α-chlordane, oxychlordane, cis-nonachlor, trans-nonachlor, and nonachlor III. A total of 10 individual PBDE congeners were measured (IUPAC numbers 28, 47, 49, 66, 85, 99, 100, 153, 154, and 183). In body samples, the limits of quantitation (LOQs) ranged from less than 0.059 ng/g wet weight to less than 0.34 ng/g wet weight for individual PCB congeners; from less than 0.23 ng/g wet weight to less than 0.35 ng/g wet weight for DDTs, aldrin, dieldrin, chlordanes, mirex, and HCHs; from less than 0.64 ng/g wet weight to less than 0.93 ng/g wet weight for endosulfan I; and from less than 0.28 ng/g wet weight to less than 0.34 ng/g wet weight for HCB.

Summed ( $\Sigma$ ) PCBs ( $\Sigma$ PCBs) were calculated by adding the concentrations of 17 commonly detected chlorobiphenyl congeners (IUPAC numbers 18, 28, 44, 52, 95, 101, 105, 118, 128, 138, 153, 170, 180, 187, 195, 206, and 209) and multiplying the result by two. This formula provides a good estimate of the total PCBs in a typical environmental sample of sediments or animals feeding on lower trophic levels (Lauenstein et al. 1993). The  $\Sigma$ DDTs were calculated by summing

the concentrations of p,p'-DDT, p,p'-DDE, p,p'-DDD, o,p'-DDD, o,p'-DDD, and o,p'-DDT. Summed chlordanes were determined by adding the concentrations of heptachlor, heptachlor epoxide,  $\gamma$ -chlordane,  $\alpha$ -chlordane, oxychlordane, cis-nonachlor, trans-nonachlor, and nonachlor III. The  $\Sigma$ HCHs were calculated by adding the concentrations of  $\alpha$ -HCH,  $\beta$ -HCH, and lindane ( $\gamma$ -HCH). The  $\Sigma$ PBDEs were calculated by adding the concentrations of the 10 PBDE congeners measured.

In addition to PBDEs and OC contaminants, feed and body samples were analyzed for low (2-3-ring) and high (4-6-ring) molecular weight PAHs using capillary column GC-MS (Sloan et al. 2005). Summed low-molecular-weight aromatic hydrocarbons (ΣLAHs) were determined by adding the concentrations of biphenyl, naphthalene, 1-methylnaphthalene, 2-methylnaphthalene, 2,6-dimethylnapthalene, acenaphthylene, acenaphthene, 2,3,5-trimethylnaphthalene, fluorene, dibenzothiophene, phenanthrene (PHN), 1-methylphenanthrene, and anthracene. Summed high-molecular-weight aromatic hydrocarbons (ΣHAHs) were calculated by adding the concentrations of fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[a]pyrene (BaP), benzo[e]pyrene, perylene, dibenz[a,h]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, indenopyrene, and benzo[g,h,i]perylene. The  $\Sigma PAHs$  were calculated by adding  $\Sigma$ HAHs and  $\Sigma$ LAHs. The LOQs for individual PAHs ranged from less than 0.13 ng/g wet weight to less than 0.47 ng/g wet weight in Chinook salmon body samples and from less than 0.087 to 0.37 ng/g wet weight in food samples.

To monitor the accuracy of the GC–MS method, a National Institute of Standards and Technology (NIST) Standard Reference Material (SRM) blue mussel Mytilus edulis homogenate (NIST SRM 1974b) and a fish tissue homogenate (NIST SRM 1947) were analyzed with each sample set and the results met laboratory criteria (Sloan et al. 2006). One (12.5%) out of eight feed samples was analyzed in duplicate to measure the precision of the method, and the laboratory quality assurance (QA) criteria were met for all analytes measured in the feed samples. Method blanks also met laboratory criteria. The QA procedures and criteria are described in detail by Sloan et al. (2006). The percent recoveries of the surrogate standards ranged from 75% to 106%.

To adjust for the influence of lipid on toxicity, we normalized body contaminant concentrations for lipid and relied primarily on lipid-normalized data to evaluate potential health effects of toxicants on juvenile salmon. Wet-weight data are also presented to facilitate comparison with other studies and to evaluate risks to

predators that consume salmon with accumulated toxicants.

Polycyclic aromatic hydrocarbon metabolites in Chinook salmon bile.—Due to the relatively small volume of bile that can be collected from individual subyearling Chinook salmon, bile samples were composited from 30 individual fish per hatchery to provide an adequate sample volume (>25 μL) for high-performance liquid chromatography (HPLC)fluorescence analysis. No PAH metabolite data were acquired for fish from Little White Salmon NFH because the volume of the bile composite was too small (<25 μL). Bile samples were analyzed for metabolites of PAHs using a HPLC-fluorescence detection method described by Krahn et al. (1984). Briefly, bile was injected directly onto a C-18 reverse-phase column (Phenomenex Synergi Hydro) and eluted with a linear gradient from 100% water (containing a trace amount of acetic acid) to 100% methanol at a flow of 1.0 mL/ min. Chromatograms were recorded at the following wavelength pairs: (1) 260–380 nm, where several 3–4ring compounds (e.g., PHN) fluoresce; and (2) 380-430 nm, where 4-5-ring compounds (e.g., BaP) fluoresce. Peaks eluting after 5 min were integrated, and the areas of these peaks were summed. The concentrations of fluorescent PAHs in the bile samples of juvenile fall Chinook salmon were determined using PHN and BaP as external standards and converting the fluorescence response of bile to PHN (ng PHN equivalents/g bile) and BaP (ng BaP equivalents/g bile) equivalents.

To ensure that the HPLC-fluorescence system was operating properly, a PHN-BaP calibration standard was analyzed at least five times, and a relative SD of less than 10% was obtained for each PAH. As part of our laboratory QA plan, two QA samples (a method blank and a fish bile control sample [bile of Atlantic salmon *Salmo salar* exposed to Monterey crude oil at  $25~\mu g/mL$  for 48~h]) were analyzed with the fish bile samples (Sloan et al. 2006).

Biliary protein was measured according to the method described by Lowry et al. (1951). Biliary fluorescence values were normalized to protein content, which is an indication of feeding state and water content of the bile. Fish that have not eaten for several days exhibit higher biliary fluorescent aromatic compound values and higher protein content than fish that are feeding constantly and excreting bile more frequently (Collier and Varanasi 1991).

Fish condition factor.—To provide a measure of fish weight adjusted for size, Fulton's condition factor (Ricker 1975) was calculated for hatchery fish as [(gutted body weight, g)/(fork length, mm)<sup>3</sup>]  $\times$  100.

Estimation of hatchery contribution to in-river fall Chinook salmon body burdens.—To estimate the relative proportions of contaminants absorbed during hatchery rearing as compared with the proportions absorbed while fish were in the lower Columbia River, concentrations of PCBs, DDTs, and PBDEs in the hatchery fall Chinook salmon were compared with concentrations of these contaminants in composite body samples (carcasses and internal organs minus stomach contents) of juvenile fall Chinook salmon of hatchery origin collected in the lower Columbia River in 2005. This collection was part of a larger cooperative project conducted in collaboration with the U.S. Geological Survey, the lower Columbia River Estuary Partnership, and the Bonneville Power Administration (LCREP 2007). The subset of samples used for these calculations consisted only of marked (i.e., adipose-fin clipped) hatchery fall Chinook salmon from lower Columbia River stocks, so their contaminant body burdens are representative of fish that are released from lower Columbia River hatcheries and subsequently spend some time rearing and migrating in the river. However, the specific hatcheries from which the fish were released are unknown. In-river fish samples came from three sites: Warrendale, the Confluence, and Columbia City. The Warrendale site (45°36'45"N, 122°01'35"W) is located in a rural forested area at river kilometer (rkm) 227, near the town of Warrendale, Oregon, just downstream from Bonneville Dam. The Confluence site (45°38′27″N, 122°43′08″W) is located at rkm 163, just downstream from the major urban centers of Portland, Oregon, and Vancouver, Washington. The Columbia City site (46°09′96″N, 122°94′51″W) is located at rkm 134 in a rural area where forestry and agriculture are the primary land uses. However, this site is affected by municipal wastewater discharges and discharges from local industries. Additional information about the sites and the sampling and analysis procedures is given by the Lower Columbia River Estuary Partnership (LCREP 2007).

Estimated PCB, DDT, and PBDE contributions from the hatchery to fish body burdens for each site were calculated as described by Meador et al. (2002). For each contaminant, the first step in this analysis was to determine the total amount, in nanograms, for each source (i.e., the hatcheries and each field site). The data from all sampled hatcheries were averaged to calculate mean hatchery fish weight and contaminant concentrations. Totals for each source were calculated as follows:

$$\begin{aligned} Total(ng) &= (mean \ fish \ weight, g) \\ &\times (concentration, ng/g \ wet \ weight). \end{aligned}$$

The contributions from river exposure and hatchery exposure (in ng/g wet weight) to the contaminant concentrations in the samples from each source were then calculated:

$$Contribution_{river} = \frac{total_{field \ site} - total_{hatchery}}{mean \ fish \ weight_{field \ site}},$$

$$Contribution_{hatchery} = \frac{total_{hatchery}}{mean \ fish \ weight_{field \ site}}.$$

Finally, the percent hatchery contribution for each sample source was calculated as

Hatchery contribution (%) = 
$$\frac{\text{contribution}_{\text{hatchery}}}{\text{concentration}_{\text{source}}} \times 100.$$

Statistical analyses.—Analysis of variance (AN-OVA) and Tukey's multiple range tests were used to identify differences in length, weight, and condition factor among fish from different hatcheries and differences in contaminant concentrations among feed samples obtained from different suppliers (Zar 1984; Dowdy and Wearden 1991). Linear regression analysis (Zar 1984) was used to evaluate relationships between contaminant concentrations in hatchery feed, contaminant concentrations in Chinook salmon bodies, and PAH metabolites in bile. Regression analysis was also used to examine the correlation between lipid content values obtained through gravimetric analysis and those obtained through TLC-FID analysis. Before statistical analyses, data were normalized through log-transformation as necessary. In addition, data on contaminant concentrations in feed and fish bodies were used to calculate the percentage of dietary contaminant present in fish bodies.

#### Results

Fish Size and Condition

Mean length of juvenile fall Chinook salmon (Table 1) differed significantly among the hatcheries, with values ranging from 67 mm at Little White Salmon NFH to 86 mm at Spring Creek NFH (ANOVA: P < 0.05). Similarly, fish weight (Table 1) was lowest in Chinook salmon collected at Little White Salmon NFH (mean = 3.2 g), was highest at Spring Creek NFH (mean = 6.2 g), and differed significantly among all hatcheries. Condition factor (Table 1), on the other hand, was lowest in fish from Washougal Hatchery (mean = 0.91) and highest in fish from Big Creek and Little White Salmon hatcheries (mean = 1.04).

#### Lipid Content and Lipid Classes

Feed samples.—Lipid concentrations in hatchery feed samples ranged from 12% to 22% as determined

Table 1.—Mean ( $\pm$ SE) length, weight, and condition factor of juvenile fall Chinook salmon sampled from Columbia River hatcheries (Figure 1) in 2005. Values followed by letters not in common are significantly different (analysis of variance and Tukey's multiple range test: P < 0.05).

Hatchery	n	Length (mm)	Weight (g)	Condition factor
Big Creek	10	84 ± 0.7 zy	6.1 ± 0.2 z	1.04 ± 0.012 z
Elochoman	10	$80 \pm 0.7 \text{ xw}$	$5.0 \pm 01 \text{ y}$	$0.99 \pm 0.0012 \text{ yx}$
Cowlitz	10	$78 \pm 0.8 \text{ xw}$	$5.0 \pm 0.1 \text{ yx}$	$0.99 \pm 0.0014 \text{ zyx}$
Washougal	10	$76 \pm 0.7 \text{ w}$	$4.1 \pm 0.1 \text{ xw}$	$0.91 \pm 0.0014 \text{ v}$
Little White Salmon	10	$67 \pm 0.4 \text{ v}$	$3.2 \pm 0.06 \text{ v}$	$1.04 \pm 0.014 \text{ yx}$
Spring Creek	10	$86 \pm 1.1 z$	$6.2 \pm 0.3 z$	$0.95 \pm 0.014 \text{ xwv}$
Klickitat	10	$71 \pm 0.7 \text{ v}$	$3.3 \pm 0.1 \text{ wv}$	$0.93 \pm 0.0014 \text{ wv}$
Priest Rapids	10	$81 \pm 1.3 \text{ yx}$	$5.3 \pm 0.3 \text{ y}$	$0.98 \pm 0.0014 \text{ xw}$

gravimetrically and from 8.8% to 19% as determined by TLC-FID (Table 2). On average, values determined gravimetrically were about 29% higher than values determined with TLC-FID, but the values were significantly and positively correlated ( $r^2 = 0.90$ , P = 0.001, n = 7). Lipid content was highest in feed from Klickitat Hatchery and lowest in feed from Priest Rapids Hatchery. Triglycerides were the predominant class of lipids in most of the feed samples, accounting for 68-92% of total lipids in feed from all hatcheries except Cowlitz Hatchery. At Cowlitz Hatchery, triglycerides accounted for only 44% of total lipids, while phospholipids accounted for 53% of total lipids. In feeds from the other hatcheries, phospholipids accounted for 2.8-14% of total lipids. Cholesterol

and free fatty acids were also present in feed samples in lower proportions, typically 1.4–3.5% of total lipids for cholesterol and 1.5–8.4% of total lipids for free fatty acids. The feed from the Little White Salmon NFH was unusual compared with feed from the other hatcheries, as it had a free fatty acid content of 23%.

Chinook salmon bodies.—Lipid concentrations in Chinook salmon body composites ranged from 2.6% to 6.2% as determined gravimetrically and from 2.5% to 4.8% as determined with TLC-FID (Table 2). On average, values determined gravimetrically were about 20% higher than those determined with TLC-FID, but the values were significantly and positively correlated  $(r^2 = 0.81, P = 0.002, n = 8)$ . Free fatty acids and triglycerides were the predominant lipid classes present

TABLE 2.—Mean percent lipid content (determined gravimetrically and by thin-layer chromatography—flame ionization detection [TLC–FID] as described in Ylitalo et al. 2005) and lipid classes of juvenile fall Chinook salmon and feed sampled from Columbia River hatcheries (Figure 1) in 2005. Lipid determinations were made on composite samples of 10 fish/composite (NM = not measured).

Hatchery	Gravimetric lipid content (%)	TLC-FID lipid content (%)	Wax esters— sterol esters (%)	Triglycerides (%)	Free fatty acids (%)	Cholesterol (%)	Phospholipids and other polar lipids (%)
			Chinook salmo	on bodies			
Big Creek	5.1	4.1	0.0	91.5	2.6	3.6	2.4
Elochoman	5.3	4.8	1.1	39.8	38.9	9.0	11.2
Cowlitz	4.7	3.6	1.1	32.6	51.3	7.9	7.2
Washougal	4.1	3.3	0.0	91.2	3.6	4.2	1.0
Little White Salmon	4.3	3.1	1.0	33.4	44.6	7.2	13.8
Spring Creek	2.6	2.5	0.87	35.9	48.1	8.2	6.9
Klickitat	6.2	4.7	0.90	35.6	27.0	9.7	26.8
Priest Rapids	5.1	4.7	0.91	31.5	51.8	7.3	8.5
All hatcheries (±SD)	$4.7 \pm 1.1$	$3.9 \pm 0.84$	$0.74 \pm 0.46$	$49 \pm 26$	$33.5 \pm 20.4$	$7.1 \pm 2.2$	$9.7 \pm 8.1$
	(n = 8)	(n = 8)	(n = 8)	(n = 8)	(n = 8)	(n = 8)	(n = 8)
			Fish fo	od			
Big Creek	NM	10	0	81	6.3	2.8	13.7
Elochoman	18	14	0.25	88	3.2	2.8	5.4
Cowlitz	20	17	0	44	1.5	1.4	52.7
Washougal	20	14	0	92	2.5	2.9	2.8
Little White Salmon	16	12	0	68	23	1.8	6.7
Spring Creek	18	15	0	76	7.4	2.2	14.0
Klickitat	22	19	0	88	2.5	1.7	7.8
Priest Rapids	12	8.8	0	79	8.4	3.5	9.4
All hatcheries (±SD)	$18 \pm 3.2$	$14 \pm 3.4$	$0.031 \pm 0.090$	$77 \pm 15$	$6.9 \pm 7.1$	$2.4 \pm 0.72$	$14 \pm 16$
	(n = 7)	(n = 8)	(n = 8)	(n = 8)	(n = 8)	(n = 8)	(n = 8)

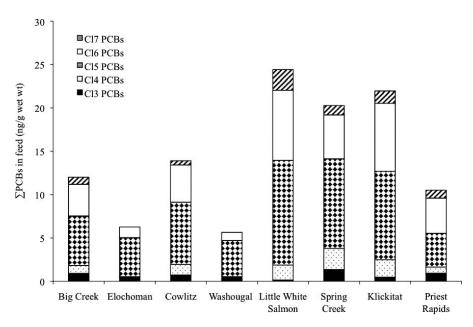


FIGURE 2.—Concentrations (ng/g wet weight) of summed ( $\Sigma$ ) PCBs measured in fish food samples obtained from Columbia River hatcheries in 2005 (Cl3, Cl4, Cl5, Cl6, and Cl7 PCBs = PCB homologues with three, four, five, six, and seven chlorines, respectively).

in Chinook salmon bodies. Triglycerides accounted for 32–92% of total lipids in fish from the individual hatcheries, while free fatty acids accounted for 2.6–52% of total lipids. Cholesterol accounted for 3.6–9.7% of total lipids, while phospholipids accounted for 1.0–27% of total lipids. While statistical comparisons could not be made because only one composite sample was made per hatchery, the data on lipid classes given in Table 2 suggest that there might be differences among hatcheries in lipid profiles.

#### Contaminant Concentrations

Persistent organic pollutants in hatchery feed.—Measurable concentrations of DDTs, PCBs, and PAHs were found in feed from all eight hatcheries. Concentrations of ΣPCBs (Figure 2) ranged from 5.3 ng/g wet weight in feed from Washougal Hatchery to 25 ng/g wet weight in feed from Little White Salmon NFH. The predominant PCBs measured in feed were the pentachlorobiphenyls and hexachlorobiphenyls (e.g., PCBs 118, 138, and 153), with concentrations ranging from 0.23 to 3.3 ng/g wet weight (Figure 2). Of the PCB congeners, 13 congeners (PCBs 156, 158, 171, 177, 191, 194, 195, 199, 205, 206, 208, and 209) were less than the LOQs (<0.054–0.39 ng/g wet weight, depending on the congener and sample size) in feed samples from all hatcheries.

Concentrations of  $\Sigma DDTs$  in feed samples ranged from 9.8 ng/g wet weight in feed from Elochoman

Hatchery to 39 ng/g wet weight in feed from Cowlitz Hatchery (Figure 3). In feed, p,p'-DDE and p,p'-DDD were the predominant DDTs, accounting for about 80% and 15% of  $\Sigma$ DDTs, respectively. In all feed samples, p,p'-DDT was also present, accounting for up to 5% of total DDTs. In addition to the p,p'-substituted isomers, low concentrations of o,p'-substituted DDTs were measured in most feed samples. In feed from all hatcheries, o,p'-DDD was measured; o,p'-DDE was measured in feed from Big Creek, Cowlitz, Klickitat, Little White Salmon, and Priest Rapids hatcheries, while o,p'-DDT was measured only in feed from Little White Salmon NFH and Klickitat Hatchery.

Concentrations of  $\Sigma$ PAHs ranged from 100 ng/g wet weight in feed from Washougal Hatchery to 610 ng/g wet weight in feed from Big Creek Hatchery (Figure 4). Of the HAHs measured in feed, fluoranthene, pyrene, and chrysene predominated, accounting for 60–80% of  $\Sigma$ HAHs in all samples (Figure 4A). Of the LAHs measured, dimethylnaphthalene, trimethylnaphthalene, and PHN predominated in feed, making up 60–80% of  $\Sigma$ LAHs. Overall, LAHs made up 90% or more of total PAHs in feed samples (Figure 4B).

Feed samples from the hatcheries contained low concentrations of several OC pesticides and PBDEs (Table 3). Chlordanes were measured in feed at all hatcheries except Priest Rapids Hatchery, with concentrations ranging from 0.69 to 4.6 ng/g wet weight. Concentrations of HCHs were less than LOQs in feed

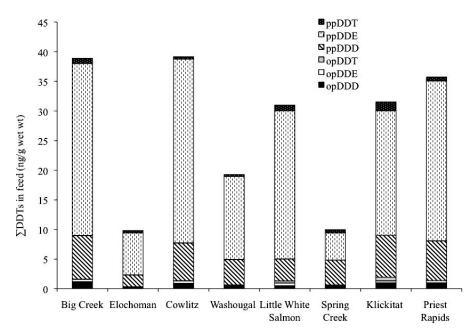


FIGURE 3.—Concentrations (ng/g wet weight) of DDT isomers and summed ( $\Sigma$ ) DDTs measured in fish food samples obtained from Columbia River hatcheries in 2005.

from both Spring Creek NFH and Washougal Hatchery, whereas the levels ranged from 0.49 to 3.2 ng/g wet weight in feed from the other hatcheries. Dieldrin and HCB were measured in feed from some of the hatcheries, but the concentrations were less than 2 ng/g wet weight in all cases. Concentrations of mirex, aldrin, and endosulfan I were below LOQs at all hatcheries. In addition, concentrations of  $\Sigma PBDEs$  were low (<3 ng/ g wet weight) in all feed samples. While statistical comparisons could not be made because only one feed sample was analyzed per hatchery, the POP concentrations in the samples we analyzed suggest there may be some differences in concentrations of DDTs, PAHs, and PCBs in feeds from the different hatcheries; concentrations of other contaminants were uniformly low.

Concentrations of contaminants were generally similar in feed samples obtained from the three different suppliers used by the hatcheries. Of the eight samples analyzed, four were from one supplier, three were from another supplier, and one was from a third supplier. Mean concentrations of  $\Sigma$ PCBs ranged from 23 to 31 ng/g wet weight, mean concentrations of  $\Sigma$ DDTs ranged from 7.8 to 22 ng/g wet weight, mean concentrations of  $\Sigma$ PBDEs ranged from 4.2 to 8.0 ng/g wet weight, and mean concentrations of  $\Sigma$ PAHs ranged from 150 to 480 ng/g wet weight. Concentrations were not significantly different among the suppliers for any

of the contaminants measured (ANOVA: 0.10 < P < 0.80).

Persistent organic pollutants in Chinook salmon bodies.—The primary contaminants found in Chinook salmon bodies were PCBs and DDTs. Concentrations (wet-weight basis) of ΣPCBs (Figure 5A) ranged from 7.3 ng/g wet weight in fish from Cowlitz Hatchery to 58 ng/g wet weight in fish from Priest Rapids Hatchery. On a lipid-weight basis (Figure 5B), body ΣPCB concentrations in Chinook salmon ranged from 170 ng/g lipid in fish from Washougal Hatchery to 1,200 ng/g lipid in fish from Priest Rapids Hatchery.

The bodies of hatchery Chinook salmon contained a wide range of PCB congeners (Figure 5), with pentachlorobiphenyls (Cl5 congeners) and hexachlorobiphenyls (Cl6) contributing greater than 65% to ΣPCBs. For example, PCBs 101, 110, 118, 138, and 153 were measured in all samples at concentrations ranging from 0.5 to 5.6 ng/g wet weight. Four congeners (PCBs 74, 128, 180, and 158) were found in fish from Priest Rapids only, while two congeners (PCBs 18 and 33) were found only in fish from Spring Creek NFH. The PCBs 156, 170, 171, 177, 183, 191, 194, 195, 199, 205, 206, 208, and 209 were below LOQs (generally <0.2–0.3 ng/g wet weight) in all samples analyzed in the present study.

Mean wet-weight concentrations of  $\Sigma$ DDTs (Figure 6A) ranged from 4.8 ng/g in bodies of Chinook salmon from Elochoman Hatchery to 15 ng/g in bodies of

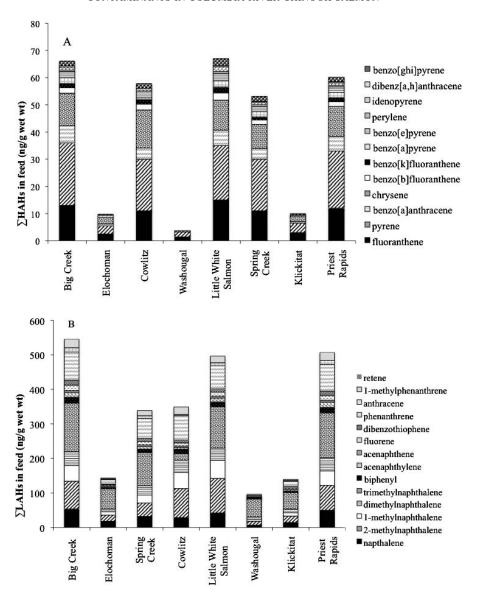


FIGURE 4.—Concentrations (ng/g wet weight) of (**A**) summed high-molecular-weight aromatic hydrocarbons (ΣHAHs) and (**B**) summed low-molecular-weight aromatic hydrocarbons (ΣLAHs) measured in fish food samples obtained from Columbia River hatcheries in 2005.

Chinook salmon from Priest Rapids Hatchery. When compared on a lipid-weight basis (Figure 6B), body  $\Sigma$ DDT concentrations ranged from 90 ng/g lipid in fish from Elochoman Hatchery to 380 ng/g lipid in fish from Big Creek Hatchery. Of the DDTs measured in Chinook salmon bodies, p,p'-DDE predominated, accounting for 75% or more of  $\Sigma$ DDTs measured. The second most prominent DDT was p,p'-DDD, which accounted for about 10–20% of DDTs measured. Concentrations of o,p'-DDD, o,p'-DDE, o,p'-DDE, o,p'-DDD, o,p'-DDE, o,p'-DDE, o,p'-DDD, o,p'-DDE, o,p'-DDE, o,p'-DDD, o,p'-DDE, o,p'-DDE, o,p'-DDE, o,p'-DDD, o,p'-DDE, o,p'-DDE, o,p'-DDE, o,p'-DDE, o,p'-DDE, o,p'-DDD, o,p'-DDE, o,p'-

DDT, and p.p'-DDT were generally low (<0.3 ng/g wet weight), accounting for 5% or less of the  $\Sigma$ DDTs in samples where they were measured.

In juvenile Chinook salmon bodies, levels of  $\Sigma$ PAHs ranged from 19 ng/g wet weight in fish from Washougal Hatchery to 42 ng/g wet weight in fish from Priest Rapids Hatchery (Figure 7). Concentrations of both LAHs and HAHs were measured, but HAH levels were below LOQs in most samples (Figure 7). Of the HAHs measured, fluoranthene was the most

common, with concentrations ranging from 0.35 ng/g in fish from Little White Salmon NFH to 0.58 ng/g in fish from Washougal Hatchery (Figure 7); other HAHs present included pyrene, benzo[e]pyrene, idenopyrene, and benzo[g,h,i]perylene. Concentrations of  $\Sigma$ LAHs in Chinook salmon bodies ranged from 16 ng/g wet weight in fish from Washougal Hatchery to 41 ng/g wet weight in fish from Priest Rapids Hatchery (Figure 7). Trimethylnaphthalene, dimethylnaphthalene, and naphthalene were the most common LAHs in bodies of Chinook salmon from most of the hatcheries.

In addition to DDTs, PCBs, and PAHs, OC pesticides and PBDEs were also measured at low concentrations in bodies of juvenile Chinook salmon from some of the hatcheries (Table 3). Chlordanes were measured in fish from all hatcheries except Cowlitz Hatchery; chlordane concentrations ranged from 0.38 to 3.0 ng/g wet weight, with highest levels found in fish from Big Creek Hatchery. Hexachlorobenzene was measured in fish from all hatcheries except Priest Rapids Hatchery and Spring Creek NFH, but concentrations were at or below 0.45 ng/g wet weight in all samples. Dieldrin, HCHs, and PBDEs were measured in fish from a few hatcheries, but concentrations were less than 1 ng/g wet weight in all cases. Mirex, aldrin, and endosulfan I were below LOQs in fish from all hatcheries.

Polycyclic aromatic hydrocarbon metabolites in Chinook salmon bile.—Levels of PAH metabolites in bile of hatchery fish ranged from 590 to 5,500 ng PHN equivalents/mg bile protein for LAH compounds fluorescing at PHN wavelengths and ranged from 24 to 210 BaP equivalents/mg bile protein for HAH compounds fluorescing at BaP wavelengths (Table 4). In fish from most hatcheries, levels of PHN metabolites were below 1,000 ng/mg bile protein and levels of BaP metabolites were below 100 ng/mg bile protein; only fish from Priest Rapids and Cowlitz hatcheries had higher levels.

Contaminant Concentrations in Feed versus Chinook Salmon Bodies and Bile

Although relationships between contaminant concentrations in feed and bodies tended to be positive, there were no statistically significant correlations between concentrations of  $\Sigma PCBs$ ,  $\Sigma DDTs$ , or  $\Sigma HAHs$  in feed and concentrations of the compounds in Chinook salmon bodies ( $\Sigma PCBs$ :  $r^2 = 0.064$ , P = 0.54, n = 8;  $\Sigma DDTs$ :  $r^2 = 0.02$ , P = 0.73, n = 8;  $\Sigma HAHs$ :  $r^2 = 0.05$ , P = 0.60, n = 8). However,  $\Sigma LAHs$  in feed and fish bodies were significantly and positively correlated ( $r^2 = 0.86$ , P = 0.0009, n = 8). Neither mean fish length ( $r^2 = 0.19$ , P = 0.28, n = 8) nor mean body lipid content ( $r^2 = 0.07$ , P = 0.50, n = 8) were significantly correlated with body burdens of

Table 3.—Concentrations (ng/g wet weight) of organochlorine pesticides and polybrominated diphenyl ethers (PBDEs) determined by gas chromatography—mass spectrometry in feed and bodies of juvenile fall Chinook salmon sampled from Columbia River hatcheries in 2005 (<LOQ = less than lower limit of quantitation; ΣHCHs = summed hexachlorocyclohexanes; ΣCHLDs = summed chlordanes; HCB = hexachlorobenzene; ΣPBDEs = summed PBDEs). Body samples are composite samples of 10 fish/composite.

Hatchery	$\Sigma H C H s^a$	Dieldrin	$\Sigma \text{CHLDs}^b$	HCB	ΣPBDEs					
Fish food (ng/g wet weight)										
Big Creek	3.2	0.20	3.3	0.49	0.95					
Elochoman	0.49	0.52	0.82	<loq< td=""><td><loq< td=""></loq<></td></loq<>	<loq< td=""></loq<>					
Cowlitz	2.0	<loq< td=""><td>1.9</td><td>0.63</td><td>1.0</td></loq<>	1.9	0.63	1.0					
Washougal	<loq< td=""><td><loq< td=""><td>0.69</td><td>0.26</td><td><loq< td=""></loq<></td></loq<></td></loq<>	<loq< td=""><td>0.69</td><td>0.26</td><td><loq< td=""></loq<></td></loq<>	0.69	0.26	<loq< td=""></loq<>					
Little White Salmon	1.6	<loq< td=""><td>4.6</td><td>1.1</td><td>2.9</td></loq<>	4.6	1.1	2.9					
Spring Creek	<loq< td=""><td>2.0</td><td>3.8</td><td>0.32</td><td>2.6</td></loq<>	2.0	3.8	0.32	2.6					
Klickitat	0.52	1.4	2.8	0.9	0.79					
Priest Rapids	0.98	<loq< td=""><td><loq< td=""><td>0.45</td><td>0.95</td></loq<></td></loq<>	<loq< td=""><td>0.45</td><td>0.95</td></loq<>	0.45	0.95					
Chinool	k salmon	bodies (r	ng/g wet we	eight)						
Big Creek	0.46	0.19	3.0	0.34	0.78					
Elochoman	<loq< td=""><td><loq< td=""><td>1.1</td><td>0.26</td><td><loq< td=""></loq<></td></loq<></td></loq<>	<loq< td=""><td>1.1</td><td>0.26</td><td><loq< td=""></loq<></td></loq<>	1.1	0.26	<loq< td=""></loq<>					
Cowlitz	<loq< td=""><td><loq< td=""><td><loq< td=""><td>0.34</td><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<>	<loq< td=""><td><loq< td=""><td>0.34</td><td><loq< td=""></loq<></td></loq<></td></loq<>	<loq< td=""><td>0.34</td><td><loq< td=""></loq<></td></loq<>	0.34	<loq< td=""></loq<>					
Washougal	<loq< td=""><td><loq< td=""><td>1.7</td><td>0.28</td><td><loq< td=""></loq<></td></loq<></td></loq<>	<loq< td=""><td>1.7</td><td>0.28</td><td><loq< td=""></loq<></td></loq<>	1.7	0.28	<loq< td=""></loq<>					
Little White Salmon	<loq< td=""><td><loq< td=""><td>1.1</td><td>0.31</td><td><loq< td=""></loq<></td></loq<></td></loq<>	<loq< td=""><td>1.1</td><td>0.31</td><td><loq< td=""></loq<></td></loq<>	1.1	0.31	<loq< td=""></loq<>					
Spring Creek	<loq< td=""><td>0.37</td><td>0.38</td><td><loq< td=""><td>0.71</td></loq<></td></loq<>	0.37	0.38	<loq< td=""><td>0.71</td></loq<>	0.71					
Klickitat	<loq< td=""><td>0.39</td><td>1.1</td><td>0.45</td><td><loq< td=""></loq<></td></loq<>	0.39	1.1	0.45	<loq< td=""></loq<>					
Priest Rapids	<loq< td=""><td><loq< td=""><td>0.46</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<>	<loq< td=""><td>0.46</td><td><loq< td=""><td><loq< td=""></loq<></td></loq<></td></loq<>	0.46	<loq< td=""><td><loq< td=""></loq<></td></loq<>	<loq< td=""></loq<>					

 $<sup>^</sup>a$  Includes  $\alpha\text{-HCH},$   $\beta\text{-HCH},$  and  $\gamma\text{-HCH}$  (lindane).

PCB, DDTs, or PAHs. Concentrations of LAHs in feed showed significant correlations with levels of PAH metabolites in bile fluorescing at PHN wavelengths (an estimator of exposure to LAHs); for LAHs in feed (ng/g wet weight) versus PHN metabolites (ng equivalents/g bile), the  $r^2$  value was 0.91 (P = 0.0008, n = 7). However, concentrations of HAHs in feed showed no correlation ( $r^2 = 0.25$ , P = 0.26, n = 7) with levels of PAH metabolites in bile fluorescing at BaP wavelengths (an estimator of exposure to HAHs).

Generally, contaminant concentrations on a lipid weight basis were similar between feed and Chinook salmon bodies or were lower in feed than in fish bodies (Table 5). Depending on the hatchery, DDT concentrations in juvenile Chinook salmon were 96–380% of DDT concentrations in feed. For the majority of hatcheries, PCB concentrations in bodies were in a similar range (110-480% of the PCB concentrations in feed). However, in fish from Elochoman, Washougal, and Priest Rapids hatcheries, PCB concentrations in fish were much higher relative to PCB concentrations in feed than was typical. In fish from Washougal and Elochoman hatcheries, body PCB concentrations were 780\% of those in feed; in fish from Priest Rapids Hatchery, body PCB concentrations were 1,300% of those in feed. Similarly, for chlordanes, juvenile

b Includes heptachlor, heptachlor epoxide, γ-chlordane, α-chlordane, oxychlordane, cis-nonachlor, trans-nonachlor, and nonachlor III.

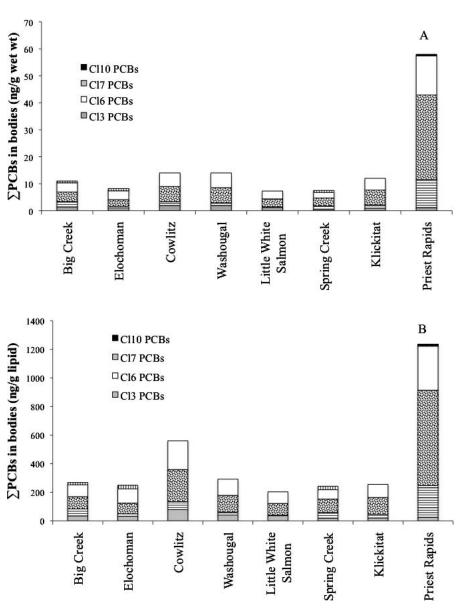


FIGURE 5.—Concentrations of summed ( $\Sigma$ ) PCBs in (A) nanograms per gram of wet weight and (B) nanograms per gram of lipid, measured in body composites (10 fish/composite) of juvenile fall Chinook salmon collected from Columbia River hatcheries in 2005 (Cl3, Cl6, Cl7, and C110 PCBs = PCB homologues with 3, 6, 7, and 10 chlorines).

Chinook salmon typically contained from 44% to 140% of the levels in feed. At Big Creek, Elochoman, and Washougal hatcheries, however, chlordane concentrations in fish bodies were 230–1,200% of those in feed. Concentrations of HCHs, dieldrin–aldrin, HCB, and PBDEs were generally too low in both fish bodies and feed for comparisons to be calculated, but where they were present, concentrations in fish bodies ranged from 36% to 540% of the concentrations in feed, with values

at Big Creek and Washougal hatcheries generally among the highest. Concentrations of LAHs and HAHs in bodies were considerably lower than those in feed. In Chinook salmon bodies, HAHs for the most part were barely detected, with maximum levels typically less than 7% of the concentrations found in feed. The exception was for fish from Washougal Hatchery, in which body HAH concentrations were 420% of the HAH concentration in feed. Concentrations of LAHs

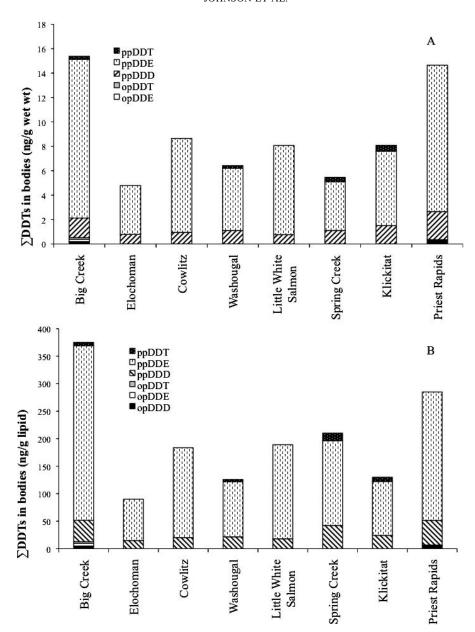


FIGURE 6.—Concentrations of DDT isomers and summed ( $\Sigma$ ) DDTs in (**A**) nanograms per gram of wet weight and (**B**) nanograms per gram of lipid, measured in body composites (10 fish/composite) of juvenile fall Chinook salmon collected from Columbia River hatcheries in 2005.

were generally higher in bodies relative to feed than was the case for HAHs; body LAH concentrations ranged from 16% to 84% of concentrations in feed.

Hatcheries as a Source of Contamination in Lower Columbia River Fall Chinook Salmon

In juvenile fall Chinook salmon of hatchery origin from the lower Columbia River, the estimated proportions of PCBs, DDTs, and PBDEs absorbed during hatchery rearing were generally lower than the proportions absorbed in the river, although this varied with the contaminant and the site where the fish were collected (Tables 6–8). For PCBs, it was estimated that Chinook salmon from Warrendale would have accumulated only 4.6 ng PCBs/g of fish after leaving the hatchery, while those collected at the Confluence

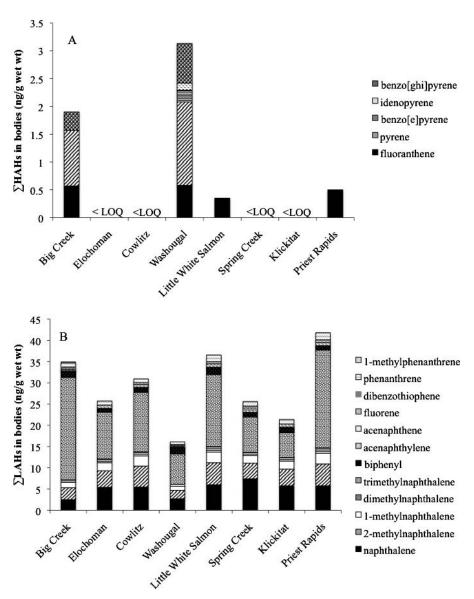


FIGURE 7.—Concentrations (ng/g wet weight) of (A) summed high-molecular-weight aromatic hydrocarbons ( $\Sigma$ HAHs) and (B) summed low-molecular-weight aromatic hydrocarbons ( $\Sigma$ LAHs) measured in body composites (10 fish/composite) of juvenile fall Chinook salmon collected from Columbia River hatcheries in 2005 (LOQ = limit of quantitation).

would have accumulated about 52 ng PCBs/g (Table 6). The estimated proportion of PCBs that could be attributed to hatchery exposure ranged from 76% in Chinook salmon from Warrendale to only 20% in fish from the Confluence. For DDTs (Table 7), it was estimated that fish from Warrendale and the Confluence would have accumulated 12–14 ng DDTs/g after leaving the hatchery, while fish collected at Columbia City would have accumulated about 30 ng DDTs/g (Table 7). The estimated proportion of DDTs that could

be attributed to hatchery exposure ranged from 20% in Chinook salmon from Columbia City to 40% in fish from Warrendale. In the case of PBDEs (Table 8), the contribution of the hatchery to body burdens was minimal in fish from all sites, ranging from less than 1% to 4% total PBDEs.

#### Discussion

Our analyses indicate that chemical contaminants are present in the feed and bodies of juvenile fall Chinook

TABLE 4.—Concentrations (ng/mg of bile protein) of metabolites of high- and low-molecular-weight polycyclic aromatic hydrocarbons (PAHs) measured in bile of juvenile fall Chinook salmon sampled from Columbia River hatcheries (Figure 1) in 2005. Low-molecular-weight metabolites were measured at phenanthrene (PHN) wavelengths, while high-molecular-weight metabolites were measured at benzo[a]py-rene (BaP) wavelengths. Each bile sample was a composite of approximately 30 fish (NM = not measured).

Hatchery	PHN <sup>a</sup> (ng equivalents/mg bile protein)	BaP <sup>b</sup> (ng equivalents/mg bile protein)
Big Creek State	590	33
Elochoman	700	27
Cowlitz	2,000	71
Washougal	920	22
Little White Salmon	NM	NM
Spring Creek	930	43
Klickitat	600	24
Priest Rapids	5,500	210

<sup>&</sup>lt;sup>a</sup> Concentrations in parts per billion (ng/g) based on total area compared with the fluorescence of the PHN standard at 260 and 380nm wavelengths.

salmon from a number of hatcheries in the Columbia River basin, although at relatively low concentrations. The most widespread contaminants were PCBs, DDTs, and LAHs, which were observed in fish and feed from all hatcheries. Some additional OC pesticides were also detected. The PBDEs, in contrast, were below LOQs in most samples.

Concentrations of POPs in feed samples were similar to or below concentrations reported in previous studies. In feed samples from U.S. Fish and Wildlife Service hatcheries, Maule et al. (2007) reported PCB concentrations in about the same range (<1-11 ng/g wet weight) as those we measured. Somewhat higher PCB

concentrations were reported in commercial feed samples analyzed by Easton et al. (2002), Hites et al. (2004a), and Kelly et al. (2008), where a number of samples contained PCBs at concentrations in the range of 30-90 ng/g wet weight. Similarly, levels of PBDEs in the fish feeds we sampled were in the lower range of values reported by other investigators (<1-11 ng/g wet weight; Hites et al. 2004b; Montory and Barra 2006). Concentrations of DDTs in most of our samples were 30-40 ng/g wet weight, which is comparable with values in the range of 3-60 ng/g wet weight reported by Easton et al. (2002), Hites et al. (2004a), Maule et al. (2007), and Kelly et al. (2008). In our feed samples, p,p'-DDE and p,p'-DDD accounted for the majority of  $\Sigma$ DDTs measured, which is similar to results of previous studies (Easton et al. 2002; Hites et al. 2004a; Maule et al. 2007). However, low proportions of o,p'-DDT and p,p'-DDT, the parent forms of the compounds, were also present in most of our feed samples, suggesting that the feed contained products (e.g., fish oils, fish meals) from regions with relatively recent usage of DDTs (Jacobs et al. 2002).

Concentrations of PAHs in the hatchery feeds were higher than expected (up to 600 ng/g wet weight). More typical concentrations are below 200 ng/g wet weight (Easton et al. 2002; Johnson et al. 2007a), although concentrations over 1,000 ng/g wet weight have occasionally been reported (Easton et al. 2002). Commercial fish feeds, which consist primarily of fish meal and fish oil (Naylor et al. 2000), usually contain negligible levels of PAHs because these compounds are metabolized and eliminated by fish and do not accumulate in their tissues (Varanasi et al. 1989; Hom et al. 1996, 1999). Although the sources of the PAHs in the fish feeds we analyzed are not known, the composition of the PAHs measured in the feed (e.g.,

Table 5.—Contaminant concentrations (%) in fish bodies versus feed for juvenile fall Chinook salmon sampled from Columbia River hatcheries (Figure 1) in 2005. Values are body concentrations expressed as percentages of concentrations in feed. Calculations are based on lipid weight concentrations for both body and feed samples ( $\Sigma$  = summed concentrations; PCBs = polychlorinated biphenyls; HCHs = hexachlorocyclohexanes; CHLDs = chlordanes; HCB = hexachlorobenzene; PBDEs = polybrominated diphenyl ethers; LAHs = low-molecular-weight aromatic hydrocarbons; HAHs = high-molecular-weight aromatic hydrocarbons; <LOQ = less than lower limit of quantitation).

Hatchery	ΣPCBs	ΣDDTs	ΣHCHs <sup>a</sup>	Dieldrin	ΣCHLDs <sup>b</sup>	НСВ	ΣPBDEs	ΣLAHs	ΣΗΑΗs
Big Creek	230	96	36	<loq< td=""><td>230</td><td>170</td><td>200</td><td>16</td><td>7.2</td></loq<>	230	170	200	16	7.2
Elochoman	780	160	<loq< td=""><td><loq< td=""><td>450</td><td><loq< td=""><td><loq< td=""><td>59</td><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<>	<loq< td=""><td>450</td><td><loq< td=""><td><loq< td=""><td>59</td><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<>	450	<loq< td=""><td><loq< td=""><td>59</td><td><loq< td=""></loq<></td></loq<></td></loq<>	<loq< td=""><td>59</td><td><loq< td=""></loq<></td></loq<>	59	<loq< td=""></loq<>
Cowlitz	210	96	<loq< td=""><td><loq< td=""><td><loq< td=""><td>230</td><td><loq< td=""><td>37</td><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<></td></loq<>	<loq< td=""><td><loq< td=""><td>230</td><td><loq< td=""><td>37</td><td><loq< td=""></loq<></td></loq<></td></loq<></td></loq<>	<loq< td=""><td>230</td><td><loq< td=""><td>37</td><td><loq< td=""></loq<></td></loq<></td></loq<>	230	<loq< td=""><td>37</td><td><loq< td=""></loq<></td></loq<>	37	<loq< td=""></loq<>
Washougal	780	170	<loq< td=""><td><loq< td=""><td>1,200</td><td>540</td><td><loq< td=""><td>84</td><td>420</td></loq<></td></loq<></td></loq<>	<loq< td=""><td>1,200</td><td>540</td><td><loq< td=""><td>84</td><td>420</td></loq<></td></loq<>	1,200	540	<loq< td=""><td>84</td><td>420</td></loq<>	84	420
Little White Salmon	110	96	<loq< td=""><td><loq< td=""><td>88</td><td>100</td><td><loq< td=""><td>28</td><td>1.9</td></loq<></td></loq<></td></loq<>	<loq< td=""><td>88</td><td>100</td><td><loq< td=""><td>28</td><td>1.9</td></loq<></td></loq<>	88	100	<loq< td=""><td>28</td><td>1.9</td></loq<>	28	1.9
Spring Creek	480	380	<loq< td=""><td>130</td><td>68</td><td><loq< td=""><td>190</td><td>53</td><td><loq< td=""></loq<></td></loq<></td></loq<>	130	68	<loq< td=""><td>190</td><td>53</td><td><loq< td=""></loq<></td></loq<>	190	53	<loq< td=""></loq<>
Klickitat	190	91	<loq< td=""><td>97</td><td>140</td><td>170</td><td><loq< td=""><td>52</td><td><loq< td=""></loq<></td></loq<></td></loq<>	97	140	170	<loq< td=""><td>52</td><td><loq< td=""></loq<></td></loq<>	52	<loq< td=""></loq<>
Priest Rapids	1,300	97	<loq< td=""><td><loq< td=""><td>44</td><td><loq< td=""><td><loq< td=""><td>19</td><td>1.9</td></loq<></td></loq<></td></loq<></td></loq<>	<loq< td=""><td>44</td><td><loq< td=""><td><loq< td=""><td>19</td><td>1.9</td></loq<></td></loq<></td></loq<>	44	<loq< td=""><td><loq< td=""><td>19</td><td>1.9</td></loq<></td></loq<>	<loq< td=""><td>19</td><td>1.9</td></loq<>	19	1.9

<sup>&</sup>lt;sup>a</sup> Includes  $\alpha$ -HCH,  $\beta$ -HCH, and  $\gamma$ -HCH (lindane).

b Concentrations in parts per billion (ng/g) based on total area compared with the fluorescence of the BaP standard at 380 and 430 nm wavelengths.

<sup>&</sup>lt;sup>b</sup> Includes heptachlor, heptachlor epoxide, g-chlordane, a-chlordane, oxychlordane, *cis*-nonachlor, *trans*-nonachlor, and nonachlor III.

Table 6.—Estimated accumulation of polychlorinated biphenyls (PCBs) in juvenile fall Chinook salmon sampled from the lower Columbia River and estuary. Fish from Warrendale, the Confluence, and Columbia City sites are marked hatchery released fish that were collected in the river in spring 2005. All fish belong to lower Columbia River stocks, but their hatcheries of origin are unknown.

Source	River kilometer	Mean weight of fish (g)	PCB concentration (ng/g)	Total PCBs (ng)	Concentration from river exposure (ng/g) <sup>c</sup>	Concentration from hatchery exposure (ng/g)	Percentage of PCBs from hatchery exposure
Warrendale <sup>a</sup>	227	5.2	19	99	4.6	14	76
Confluence <sup>a</sup>	163	5.6	65	364	52	13	21
Columbia City <sup>a</sup>	134	5.4	49	265	35	14	28
Columbia River hatcheries <sup>b</sup>		5	15	75	0	15	100

<sup>&</sup>lt;sup>a</sup> Data on body PCB concentrations from these sites are from the Lower Columbia River Estuary Partnership (LCREP 2007).

high proportions of naphthalene and alkylated naphthalenes) indicate that the feed or feed components were exposed to a petroleum product that contains these compounds, possibly at the hatcheries themselves.

The hatcheries sampled as part of this study obtained their feeds from several commercial feed suppliers based in the Pacific Northwest, so these feeds would probably be representative of those used by other trout and salmon hatcheries in the region. In fact, their contaminant concentrations were similar to those reported for feeds used by salmon hatcheries in British Columbia (Kelly et al. 2008). Our study was not designed to evaluate contaminant concentrations in feeds supplied by different companies, and the samples we collected would not be sufficient to characterize the various types of feed produced by these suppliers. However, we did find that the average concentrations of DDTs, PCBs, and PAHs were not significantly different in feeds supplied by different manufacturers.

For the most part, average body burdens of bioaccumulative POPs were similar in fish from hatcheries throughout the Columbia River area and somewhat lower than those previously reported for farmed salmon or juvenile fall Chinook salmon from other Oregon and Washington hatcheries (i.e., about 50

ng/g wet weight for PCBs and DDTs and up to 5 ng/g wet weight for PBDEs; Easton et al. 2002; Jacobs et al. 2002; Hites et al. 2004a, 2004b; Johnson et al. 2007a). In part, this is because the fish from the studies by Easton et al. (2002), Hites et al. (2004a, 2004b), and Johnson et al. (2007a) were generally older and larger than those examined in the present study and thus would have had a longer period to accumulate contaminants. Also, contaminant concentrations in feed were higher in some of the earlier studies. This appears to be a general trend; since the 1970s, levels of contaminants in fish feeds have tended to decline (Maule et al. 2007). In our study, Chinook salmon body contaminant concentrations comparable with those in fish from the studies cited above were found only in fish from Priest Rapids Hatchery, with body PCB levels that were almost 60 ng/g wet weight.

Concentrations of PCBs, DDTs, chlordanes, and other POPs measured on a lipid weight basis were generally up to 300% of concentrations in feed. However, contaminant concentrations in Chinook salmon bodies were not strongly correlated with concentrations in feed samples from the hatcheries where the fish were reared. This is perhaps not surprising, as a variety of factors that we could not

TABLE 7.—Estimated accumulation of DDTs in juvenile fall Chinook salmon from the lower Columbia River and estuary. Fish from Warrendale, the Confluence, and Columbia City sites are marked hatchery-released fish that were collected in the river in spring 2005. All fish belong to lower Columbia River stocks, but their hatcheries of origin are unknown.

Source	River kilometer	Mean weight of fish (g)	DDT concentration (ng/g)	Total DDTs (ng)	Concentration from river exposure (ng/g) <sup>c</sup>	Concentration from hatchery exposure (ng/g)	Percentage of DDTs from hatchery exposure
Warrendale <sup>a</sup>	227	5.2	20	104	12	8.1	40
Confluence <sup>a</sup>	163	5.6	22	123	15	7.5	34
Columbia City <sup>a</sup>	134	5.4	38	205	30	7.8	20
Columbia River hatcheries <sup>b</sup>		5	8.3	42	0	8.3	100

<sup>&</sup>lt;sup>a</sup> Data on body DDT concentrations from these sites are from the Lower Columbia River Estuary Partnership (LCREP 2007).

<sup>&</sup>lt;sup>b</sup> Mean of data from Elochoman, Spring Creek, Cowlitz, Little White Salmon, Klickitat, and Priest Rapids hatcheries (Figure 1).

<sup>&</sup>lt;sup>c</sup> Concentrations of PCBs in fish from the estuary were determined by subtracting total nanograms for hatchery fish from the total nanograms in estuary fish and dividing by the weight of estuary fish (see Meador et al. 2002). All concentrations are reported on a wet-weight basis.

<sup>&</sup>lt;sup>b</sup> Mean of data from Elochoman, Spring Creek, Cowlitz, Little White Salmon, Klickitat, and Priest Rapids hatcheries (Figure 1).

<sup>&</sup>lt;sup>c</sup> Concentrations of DDTs in fish from the estuary were determined by subtracting total nanograms for hatchery fish from the total nanograms in estuary fish and dividing by the weight of estuary fish (see Meador et al. 2002). All concentrations are reported on a wet-weight basis.

TABLE 8.—Estimated accumulation of polybrominated diphenyl ethers (PBDEs) in juvenile fall Chinook salmon from the lower Columbia River and estuary. Fish from Warrendale, the Confluence, and Columbia City sites are marked hatchery-released fish that were collected in the river in spring 2005. All fish belong to lower Columbia River stocks, but their hatcheries of origin are unknown.

Source	River kilometer	Mean weight of fish (g)	PBDE concentrations (ng/g)	Total PBDEs (ng)	Concentration from river exposure (ng/g) <sup>c</sup>	Concentration from hatchery exposure (ng/g)	Percentage of PBDEs from hatchery exposure
Warrendale <sup>a</sup>	227	5.2	2.9	15	2.8	0.12	4.0
Confluence <sup>a</sup>	163	5.6	8.6	48	8.5	0.11	1.2
Columbia City <sup>a</sup> Columbia River hatcheries <sup>b</sup>	134	5.4 5.0	37 0.12	200 0.6	37 0.0	0.11 0.12	0.3 100

<sup>&</sup>lt;sup>a</sup> Data on body PBDE concentrations from these sites are from the Lower Columbia River Estuary Partnership (LCREP 2007).

fully assess, including fish age, maternal transfer of contaminants, body lipid content, dietary moisture and lipid contents, contaminants in previous feeds, and feed ration, could affect absorption and body concentrations of contaminants. However, there were a few cases (e.g., ΣPCB concentrations in fish from Washougal, Elochoman, and Priest Rapids hatcheries) in which contaminant concentrations in fish bodies appeared unusually high for the dietary concentrations. These levels of contaminant uptake could not be explained by fish size or lipid content, raising the possibility that fish might be exposed to contaminants from sources other than the diet. Recently, there have been reports of fish in Montana and Washington state hatcheries being exposed to PCBs in paint (MFWP 2004; Cornwall 2005), but whether this is a possible source of exposure for the fish at the hatcheries we sampled is unknown.

In comparison with the OCs, accumulation of PAHs from feed into Chinook salmon tissues was low, with typical body concentrations of  $\Sigma$ LAHs and  $\Sigma$ HAHs below 84% and below 7.2%, respectively, of concentrations measured in feed. This is consistent with the extensive metabolism of PAHs in fish (Varanasi et al. 1989). Fish from Washougal Hatchery were an exception, as the  $\Sigma$ HAH concentration in tissues of these fish was 420% of that in feed. However, the reason for the relatively high uptake of HAHs in these fish is unknown. Concentrations of  $\Sigma$ LAHs in feed were highly correlated with levels of LAH metabolites in fish bile, indicating the importance of the diet as a source for these compounds. The relationship was not as strong for  $\Sigma$ HAHs, which were present at only low concentrations in feed samples.

In general, the concentrations of PCBs, DDTs, and PAHs measured in hatchery fish and feed in the present study were below levels associated with adverse biological effects in salmon. For PCBs, Meador et al. (2002) estimated a critical body residue of 2,400 ng/g lipid weight for protection against 95% of effects

ranging from enzyme induction to mortality. Even in Chinook salmon from Priest Rapids Hatchery, PCB body burdens were well below this level. Similarly, DDT concentrations in hatchery Chinook salmon were well below the DDT effect threshold of 600 ng/g wet weight estimated by Beckvar et al. (2005), or approximately 6,000 ng/g lipid weight if we assume that most of the data on which the threshold was based were derived from laboratory-reared salmonids, which typically have a lipid content near 10% (Meador et al. 2002). Dietary levels of PAHs were also well below concentrations that have been associated with effects on growth, metabolism, or immune function in field and laboratory studies (Casillas et al. 1998; Palm et al. 2003; Meador et al. 2006), which are generally in the range of 5,000 ng/g wet weight and above.

Levels of PAH metabolites in bile of hatchery Chinook salmon were also generally below concentrations thought to be associated with toxic effects in juvenile salmon, estimated by Meador et al. (2008) as PHN metabolite levels in excess of 2 µg/mg bile protein. Levels of PHN metabolites in bile in this range were found only in fish from Cowlitz and Priest Rapids hatcheries. According to Meador et al. (2008), dietary PAH levels typically associated with bile metabolite levels like those in Cowlitz and Priest Rapids fish are 11–22 µg/g wet weight, much higher than levels in feed from these two hatcheries. This suggests that Chinook salmon from these two hatcheries may have been exposed to PAHs from other sources, possibly through the water column.

Lipid content in Chinook salmon and feed samples was measured by both gravimetric and TLC-FID quantitation methods. Previous studies have shown that TLC-FID lipid values are generally lower than those determined gravimetrically, probably because the gravimetric method measures lipids as well as other co-extracted biogenic materials that are not included in percent lipid values determined by TLC-FID (Delbeke

<sup>&</sup>lt;sup>b</sup> Mean of data from Elochoman, Spring Creek, Cowlitz, Little White Salmon, Klickitat, and Priest Rapids hatcheries (Figure 1).

<sup>&</sup>lt;sup>c</sup> Concentrations of PBDEs in fish from the estuary were determined by subtracting total nanograms for hatchery fish from the total nanograms in estuary fish and dividing by the weight of estuary fish (see Meador et al. 2002). All concentrations are reported on a wet-weight basis.

et al. 1995). In a recent study of juvenile Chinook salmon, TLC-FID lipid content values were about 15% lower than those obtained through gravimetric analysis (Johnson et al. 2007b). Results were similar for the samples we analyzed in the present study; on average, values determined by TLC-FID were 22% lower for feed and 28% lower for fish bodies than values determined gravimetrically. However, the percent lipids determined by both methods were highly correlated, as was also demonstrated in previous studies (Delbeke et al. 1995; Ylitalo et al. 2005).

Compared with that in field-collected juvenile Chinook salmon from the lower Columbia River and other Pacific Northwest estuaries (Johnson et al. 2007a, 2007b; LCREP 2007), the lipid content of the hatchery Chinook salmon we sampled was somewhat higher (3-6% versus 1-3%). This is typical of comparisons between wild and hatchery fish (Ackman and Takeuchi 1986). The hatchery Chinook salmon also had higher proportions of free fatty acids (up to 50% of total lipids) compared with those in field-collected juvenile fall Chinook salmon (<15\% of total lipids; Johnson et al. 2007a, 2007b; LCREP 2007). This is unusual because most studies, including those with both wild and hatchery-reared salmonids, have found small amounts of free fatty acids in animal tissues (Sheridan 1989; Næsje et al. 2006; Pratoomyot et al. 2008). Diet can affect lipid profiles, including free fatty acid levels, in wild and hatchery-reared fish (Bergström 1989; dos Santos et al. 1993; Jobling and Bendiksen 2003). However, the lipid content and profile of the feeds we analyzed had no special characteristics and were similar to those reported for other feeds (Easton et al. 2002; Johnson and Barnett 2003; Hamilton et al. 2005). The implications of relatively high free fatty acid levels in hatchery fish are unclear, but high free fatty acid levels have been associated with diabetes, heart disease, and related health problems in humans and laboratory animals (Boden 2002).

Although chemical contaminants were present in Columbia River hatchery fish and feed, our analyses suggest that hatcheries are unlikely to be a major source of contaminants for most juvenile fall Chinook salmon in the lower Columbia River. In the case of PBDEs, concentrations were extremely low in both fish and feed from all of the hatcheries we sampled. Concentrations of PCBs, DDTs, and PAHs or their metabolites in hatchery fish and feed were comparable with concentrations in stomach contents, bodies, and bile of juvenile fall Chinook salmon from rural estuaries in the Pacific Northwest (Johnson et al. 2007a) but were significantly lower than concentrations measured in bodies and stomach contents of juvenile fall Chinook salmon from several sites along

the lower Columbia River (LCREP 2007). These findings were reflected in our analysis of in-river and hatchery contributions to contaminant body burdens in Columbia River out-migrant juvenile Chinook salmon of hatchery origin. The river was identified as the primary source of PBDE exposure for all juvenile fall Chinook salmon. Similarly, in fish from all sites, the river was estimated to be the greatest source of DDTs, although hatchery rearing accounted for a significant percentage of DDT uptake (20-40%). It appeared that hatchery rearing could account for most of the PCB body burden of fish from rural sites (e.g., Warrendale) where PCB contamination is minimal, but for fish rearing at or downstream from major urban centers such as Portland (i.e., fish collected from the Confluence and Columbia City sites), the river was by far a more important source of PCBs. In summary, the results suggested that high contaminant body burdens in out-migrant hatchery fall Chinook salmon are probably due to exposure to contaminants in the Columbia River rather than to rearing practices at regional hatcheries.

However, even if contaminant levels in hatchery feeds or the hatchery environment are below concentrations likely to affect fish during hatchery rearing, their accumulation in prerelease hatchery fish may be a management concern. If contaminant body burdens are already above background levels when fish leave the hatchery, they have an increased risk of reaching concentrations that could reduce their likelihood of survival during estuarine residence. Moreover, the lipid content of hatchery fish when they are released is relatively high, and when these lipids are metabolized during downstream migration, contaminants stored in body fat may also be released and exert toxic effects (Elskus et al. 2005). Finally, salmon with elevated contaminant levels would contribute toxicants to the environment and to the food chain (Missildine et al. 2005; O'Toole et al. 2006). This represents a hazard for birds and other piscivorous wildlife, which are known to be at risk from exposure to bioaccumulative contaminants (e.g., PCBs and DDTs) in the lower Columbia River (Anthony et al. 1993; Thomas and Anthony 1999; Henny et al. 2003; USFWS 2004; Buck et al. 2005.

Although contaminant concentrations were relatively low in all of the feeds we analyzed, we recommend routinely testing feeds for contaminants because other studies have shown that the types and concentrations of chemicals in feeds can vary substantially from lot to lot (Ylitalo et al. 2001; Easton et al. 2002; Hites et al. 2004a; Maule et al. 2007). The U.S. Fish and Wildlife Service already does chemical testing on feeds used in national fish hatcheries (e.g., Maule et al. 2007), and

the procedure should be encouraged for hatcheries managed by other agencies. The testing programs that have been implemented to date appear to have led to improvements in feed quality (Maule et al. 2007), and continued efforts will help to minimize any risks to fish, wildlife, and human health.

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